



The Key Role of Surface Tension in the Theory of Bubble-Assisted Explosion/Combustion Triggering

by Michael Grinfeld

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14. ABSTRACT We have revised the classical and widely accepted theory of (Bowden, F. P.; Yoffe, A. D. <i>The Initiation and Growth of Explosion in Liquids and Solids</i> ; Cambridge University Press: New York, NY, 1952) explaining the initiation of explosion/combustion by means of adiabatic compression of the dissolved gaseous bubbles within condensed liquid substances. The main thrust of our study is to explore the role of surface tension at the interface “liquid explosive/gaseous bubble.” The potential impotence of such a study is associated with smallness of the bubbles. Their smallness causes high overpressure within the bubble as compared with the pressure within the surrounding explosive/flammable liquid due to the Laplace excess pressure. It is demonstrated that the influence of the surface tension can be indeed quite dramatic.					
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1. Introduction

Triggering (initiating) is one of the key processes in combustion, explosion, and detonation of energetic and explosive materials. By controlling triggering, these strong processes can be made more probable or to be suppressed if necessary. In order to get such control, one has to understand clearly the physical nature of the triggering in different substances and environments. Depending on the substance, the effective mechanisms of the triggering can be quite different.

Explosion/combustion triggering is nothing else but initiation of corresponding chemical reactions within substances. As a rule, even relatively modest changes of temperature of the potentially explosive/combustible substances are capable of rather big accelerating combustion-like chemical reactions. The combustion is an exothermic chemical reaction accompanied by sufficiently big release of energy. Once the combustion is initially triggered, then the combustion process becomes self-supporting. It becomes self-supporting because the required sufficiently high temperature is maintained due to the energy release accompanying the combustion. Thus, the bottleneck for the combustion is the initial triggering of the reaction.

Various mechanisms of controllable triggering of explosions/combustions deserve serious further attention because of the two competing reasons. On one hand, the triggering should be easily achievable when we need the explosive/combustible materials to burn or explode. On the other hand, we have to be able to prevent these powerful processes when we do not need them, in particular, when storing them in hazardous environments. Although there are different mechanisms of triggering explosions/combustions, they all share one common feature: the reactions are usually triggered in the spatially separated hot spots, i.e., in the spots with particularly high temperatures (see figure 1).

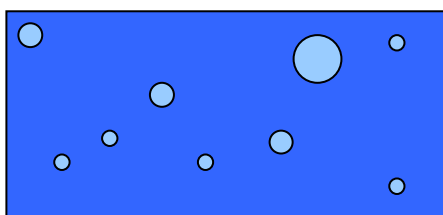


Figure 1. The gas/vapor bubbles with gas within liquid energetic substances.

Also, the mechanisms and devices of triggering the explosive and combustible materials are based on the processes which are quite different as compared with the spatio-temporal spreading of combustion itself. One of the most widespread methods of generating high-temperature spots in the explosive/combustible substances is the intensive impacting of these substances. It is well-known that short intensive impacts cause adiabatic loading of the substance. In turn,

adiabatic loading is usually accompanied with strong thermal effects, including significant raise in temperature.

The processes of triggering of hot spots due to adiabatic loading of liquid explosive/combustible materials have been explored long ago (1, 2). It was observed long ago that gaseous/vapor bubbles within condensed explosive/combustible materials can be converted into hot spots by the impact-like mechanical loading. Under adiabatic loading, the bubbles become much hotter than the surrounding condensed liquids. Thus, they are able to play the role of hot spots.

Two different mechanisms of triggering by means of gaseous/vapor bubbles should be distinguished depending of the nature of the gas inside the bubble. When the gas inside the bubble is mostly a flammable vapor of the surrounding flammable liquid, then the combustion/explosion begins *inside* the bubble—this is the in-vapor-ignition mechanism. However, if the bubble contains an inflammable, inert gas, then the explosion/combustion starts *outside* the bubble, i.e., within the flammable liquid in close proximity of the bubble—this is the in-liquid-ignition mechanism.

The currently accepted canonical analysis of the bubble-mediated triggering (1) is based on the analysis of the adiabata equation of the gas bubble alone, i.e., without the analysis of heating of the surrounding condensed phase. Of course, it was clear to the interested authors themselves that such an analysis is insufficient: after all, the rate of the chemical reactions within the condensed phases depends on the bulk temperature of these phases, not on the temperature of the immersed bubbles. However, as the starting point, the analysis (1) was mandatory and unavoidable.

The after-triggering stage of explosion/flame propagation is deeply associated with heat conduction, chemical reactions, mass, and momentum exchanges: these important processes have been intensively and carefully studied in many classical monographs and textbooks (2–4). Much less attention has been paid to the analysis of the process of triggering. This fact can create the misleading impression that the triggering processes are already quite clear and sufficiently well understood and explored. In fact, the situation is far of being so satisfactory: various aspects of triggering are far of being completely clear. For instance, the key formulas (1) of triggering by means of adiabatic compression of gaseous bubbles does not depend on the bubbles' sizes. The experiments and observations, however, are indicative of the fact that the process of triggering is size-sensitive (5, 6).

In the following section, we analyze one of the mechanisms that might be responsible for the dependence of explosions triggering upon the size of the bubbles. Namely, we take into account the surface tension acting at the boundary, separating the bubble from the surrounding condensed liquid. Such an analysis seemed to be mandatory because the bubbles are of the size of 10^{-3} cm. For this size range, the effects of the surface-tension-induced Laplace overpressure can be quite strong.

Our analysis shows that, indeed, that the bubble-based mechanism of triggering cannot be adequately developed without taking into account and understanding of the role of surface tension. The influence of the surface tension not only leads to quantitative corrections of the main formulas of Bowden and Yoffe (*I*), but it reveals the necessity of complete revision of the canonical theory of the bubble-assisted triggering of explosion/combustion.

2. The Thermodynamic Scenario of the Bubble-Generated Ignition

In this report, we analyze one of the simplest scenarios of triggering of combustion/explosion in the condensed liquid substance. Namely, we assume that the impact loading can be modeled as a sudden jump in the external pressure from the initial value p_e^0 to p_e^1 . Despite the fact that impact is a fast process, we assume that the triggering mechanism can be adequately described by the methods of classical thermodynamics alone (i.e., without the analysis of the momentum and kinetic energy exchange, as well as without any explicit modeling of heat conduction).

More specifically, we assumed that the triggering process can be split into two different events: the starting stage of adiabatic compression of the bubble and the final stage of heating of the condensed substance in the small vicinity of each bubble. The previously mentioned mechanism of the in-vapor-ignition triggering includes only the starting stage *i*, whereas the in-liquid-ignition includes both stages.

We assume that initially both—the condensed liquid and the gaseous bubble—have the same ambient temperature T^0 . Then, the pressure impact causes an adiabatic increase of the temperature within both—the liquid and the bubble. However, the temperature increase is much more pronounced within the bubble than within the liquid. This not-so-obvious assumption can be justified by the fact that the bubble's substance has much bigger compressibility than the condensed liquid. Therefore, the same pressure change is associated with much bigger mechanical work exerted on the bubble than on the liquid.* In the lowest order of approximation, we can even neglect the temperature change of the liquid during the initial stage of ignition. Thus, the starting stage of triggering is nothing else but the adiabatic compression of the gas inside compressible bubble. We suppose that the triggering event can start only if the temperature of the bubble exceeds the critical value T^* . For the in-vapor-ignition mechanism, the first stage of triggering is the only mandatory stage.

For the in-liquid-ignition mechanism to be operational, the first stage of adiabatic temperature increase inside the bubble is insufficient for the triggering since the high enough temperature should be reached not within the bubble but within the bulk of the liquid. We assume that this temperature increase can be achieved by means of heat flux at fixed pressure from the hot bubble

*For instance, pressure applied to an incompressible liquid cannot produce any work on it.

to the initially cool liquid. Thus, the final event is nothing else but the isobaric warming of the condensed substance surrounding the hot bubble. Even if the bubble is hot enough at the end of the adiabatic compression stage, its potential ability of heating the surrounding liquid can be limited because of insufficient amount of the available enthalpy inside the bubble. Thus, the bubble should be sufficiently big in size or to reach even higher temperature to carry enough enthalpy. In other words, there appears the dependence of the triggering ability upon the size of the bubble.

The full description of the second stage of triggering should be based on the sufficiently detailed analysis of the process of heat conduction in the bulk of the bubble and liquid and of the heat exchange across the liquid/bubble interface. However, in this report we limit ourselves to the simplified model. According to this model, the explosion/combustion triggering becomes possible only if the available full enthalpy in the bubble exceeds a certain critical value H^* .

Summarizing, we use the following two-elemental criterion of the explosion/combustion triggering. The absolute temperature has to exceed the critical value T^* and the total available enthalpy should exceed the critical value H^* .

The main target of our analysis is to find the critical impact overpressure p_e^1 / p_e^0 and of the overpressure dependence on the ambient temperature T_0 .

3. Analysis of the Scenario for the Case of the Ideal Gas Model

The specific internal energy e and enthalpy τ of the ideal gas are given by the following formulas:

$$e = c_v T + \text{const}, \quad (1)$$

and

$$\tau = c_p T + \text{const}, \quad (2)$$

where T is the absolute temperature and c_v and c_p are specific (constant) heat capacities at fixed volume and pressure, respectively. The caloric equation 1 of the ideal gas should be amended with the equation of state, connecting the absolute temperature with the pressure p and the specific volume v :

$$pv = RT, \quad (3)$$

where R is a constant.

The thermodynamic parameters of any two states (v_0, p_0, T_0) and (v_1, p_1, T_1) of any ideal gas, belonging to the same adiabata, are connected by the relationships:

$$p_0 v_0^\gamma = p_1 v_1^\gamma, T_0 v_0^{\gamma-1} = T_1 v_1^{\gamma-1}, T_0 p_0^{\frac{1-\gamma}{\gamma}} = T_1 p_1^{\frac{1-\gamma}{\gamma}}, \quad (4)$$

where γ is the ratio of the heat capacities: $\gamma \equiv c_p / c_v$.

The last of the formulas in equation 4 implies the relationship

$$\frac{T_1}{T_0} = \left(\frac{p_1}{p_0} \right)^{\frac{\gamma-1}{\gamma}} \rightarrow \frac{p_1}{p_0} = \left(\frac{T_1}{T_0} \right)^{\frac{\gamma}{\gamma-1}}. \quad (5)$$

Equation 5 leads to the following necessary condition of the successful first stage of triggering:

$$\frac{p_1}{p_0} \geq \left(\frac{T^*}{T_0} \right)^{\frac{\gamma}{\gamma-1}}. \quad (6)$$

The jump in the specific enthalpies between two states is given by the formula

$$\tau_1 - \tau_0 = c_p (T_1 - T_0) = c_p T_0 \left[(p_1 / p_0)^{(\gamma-1)/\gamma} - 1 \right]. \quad (7)$$

If the bubble has the initial volume V_0 , then the total accumulated enthalpy H is equal to

$$H = \frac{\gamma}{\gamma-1} \left[(p_1 / p_0)^{(\gamma-1)/\gamma} - 1 \right] p_0 V_0. \quad (8)$$

Equation 8 is implied by the following chain:

$$\begin{aligned} H &= (\tau_1 - \tau_0) V_0 / v_0 = c_p \frac{T_0 V_0}{v_0} \left[(p_1 / p_0)^{(\gamma-1)/\gamma} - 1 \right] \\ &= c_p \frac{p_0}{R} \left[(p_1 / p_0)^{(\gamma-1)/\gamma} - 1 \right] V_0 = \frac{\gamma}{\gamma-1} \left[(p_1 / p_0)^{(\gamma-1)/\gamma} - 1 \right] p_0 V_0. \end{aligned} \quad (9)$$

Using equation 8, we arrive at the second necessary condition of the in-liquid-ignition during the second stage of triggering:

$$\frac{\gamma}{\gamma-1} \left[\left(\frac{p_1}{p_0} \right)^{\frac{\gamma-1}{\gamma}} - 1 \right] \frac{p_0 V_0}{H^*} \geq 1. \quad (10)$$

In fact, the criterion (equation 10) should be treated as the condition defining the size V_0 of the bubbles in vicinity of which the in-liquid-ignition mechanism becomes possible.

4. The Role of Surface Tension in Triggering Combustion/Explosion

In the absence of the surface tension at the bubble/liquid interface, equations 6 and 10 would be sufficient for the analysis of the bubble-assisted triggering. However, these formulas link the parameters of the gas inside the bubble. We need, in fact, the criteria expressed in terms of the parameters of the liquid. The pressures inside the bubble are much bigger than the pressures inside the liquid in the case of small-size bubbles.

Let us consider a gaseous bubble inside a liquid. Because of the Laplace excess pressure, we now have to distinguish between the pressure in the bubble and in the liquid. Let σ be the surface tension at the interface “bubble-liquid.” We assume that σ is a constant, although it is known to depend on temperature. Then, let $R_0 = (3V_0/4\pi)^{1/3}$ and $R_1 = (3V_1/4\pi)^{1/3}$ be the radii of the bubble when the liquid is under the pressures of p_0^e and p_1^e , respectively. Then, because of the Laplace excess pressure under curved surfaces, the corresponding pressures p_0^b and p_1^b within the bubble are given by the following formulas:

$$p_0^b = p_0^e + \frac{2\sigma}{R_0} = p_0^e + \left(\frac{32\pi}{3}\right)^{1/3} \frac{\sigma}{V_0^{-1/3}}, \quad (11)$$

and

$$p_1^b = p_1^e + \frac{2\sigma}{R_1} = p_1^e + \left(\frac{32\pi}{3}\right)^{1/3} \frac{\sigma}{V_1^{-1/3}}. \quad (12)$$

Using the third of the adiabata relationships of equation 4, we get

$$\left(\frac{T_1}{T_0}\right)^{\frac{\gamma}{\gamma-1}} = \frac{\frac{p_1^e}{p_0^e} + \xi \frac{\sigma}{p_0^e V_0^{1/3}} \frac{V_1^{-1/3}}{V_0^{-1/3}}}{1 + \xi \frac{\sigma}{p_0^e V_0^{1/3}}}, \quad \xi \equiv \left(\frac{32\pi}{3}\right)^{1/3}, \quad (13)$$

implied by the following chain:

$$\frac{T_1}{T_0} = \left(\frac{p_1^b}{p_0^b}\right)^{\frac{\gamma-1}{\gamma}} = \left(\frac{p_1^e + 2\sigma R_1^{-1}}{p_0^e + 2\sigma R_0^{-1}}\right)^{\frac{\gamma-1}{\gamma}} = \left(\frac{p_1^e + 2\sigma V_1^{-1/3}}{p_0^e + 2\sigma V_0^{-1/3}}\right)^{\frac{\gamma-1}{\gamma}} = \left(\frac{\frac{p_1^e}{p_0^e} + \xi \frac{\sigma}{p_0^e V_0^{1/3}} \frac{V_1^{-1/3}}{V_0^{-1/3}}}{1 + \xi \frac{\sigma}{p_0^e V_0^{1/3}}}\right)^{\frac{\gamma-1}{\gamma}}. \quad (14)$$

The second of the adiabata relationships of equation 4 implies

$$\frac{V_0^{\gamma-1}}{V_1^{\gamma-1}} = \frac{T_1}{T_0} \rightarrow \frac{V_0^{-1/3}}{V_1^{-1/3}} = \left(\frac{T_1}{T_0}\right)^{-\frac{1}{3(\gamma-1)}}. \quad (15)$$

Combining equations 13 and 15, we get

$$\frac{p_1^e}{p_0^e} + \xi \frac{\sigma}{p_0^e V_0^{1/3}} \left(\frac{T_1}{T_0} \right)^{\frac{1}{3(\gamma-1)}} - \left(1 + \xi \frac{\sigma}{p_0^e V_0^{1/3}} \right) \left(\frac{T_1}{T_0} \right)^{\frac{\gamma}{\gamma-1}} = 0. \quad (16)$$

Introducing the dimensionless variables,

$$\Sigma \equiv \frac{\xi \sigma}{p_0^e V_0^{1/3}}, \quad \Pi \equiv \frac{p_1^e}{p_0^e}, \quad \Theta \equiv \frac{T_1}{T_0}, \quad (17)$$

we can rewrite equation 16 in the following dimensionless form:

$$\Pi = \Theta^{\frac{\gamma}{\gamma-1}} + \Sigma \left(\Theta^{\frac{\gamma}{\gamma-1}} - \Theta^{\frac{1}{3(\gamma-1)}} \right). \quad (18)$$

The first term in equation 18 is equivalent to the result presented by equation 6. Equation 18 can be rewritten as follows:

$$\Pi = \Theta^{\frac{\gamma}{\gamma-1}} \left[1 + \Sigma \left(1 - \frac{1}{\Theta^{\frac{1+\frac{2}{3(\gamma-1)}}}} \right) \right]. \quad (19)$$

In view of the inequalities $\gamma > 1$, $\Sigma > 0$, and $\Theta > 1$, equation 19 shows that by neglecting the surface tension, we can essentially underestimate the impact pressure which can trigger explosion/combustion.

Let us derive the correct generalization of the enthalpy condition (equation 10) for the case of non-zero surface tension. For the change of the full enthalpy, we get

$$H = \frac{\gamma}{\gamma-1} (1 + \Sigma) p_0^e V_0 (\Theta - 1), \quad (20)$$

implied by the following chain:

$$\begin{aligned} H &= \frac{V_0}{v_0} c_p (T_1 - T_0) = \frac{V_0}{v_0} c_p T_0 \left(\frac{T_1}{T_0} - 1 \right) = \frac{c_p}{R} p_0^b V_0 \left(\frac{T_1}{T_0} - 1 \right) = \frac{\gamma}{\gamma-1} \left(p_0^e + \frac{2\sigma}{R_0} \right) V_0 \left(\frac{T_1}{T_0} - 1 \right) \\ &= \frac{\gamma}{\gamma-1} \left[1 + \left(\frac{32\pi}{3} \right)^{1/3} \frac{\sigma}{p_0^e V_0^{-1/3}} \right] p_0^e V_0 \left(\frac{T_1}{T_0} - 1 \right) = \frac{\gamma}{\gamma-1} (1 + \Sigma) p_0^e V_0 \left(\frac{T_1}{T_0} - 1 \right). \end{aligned} \quad (21)$$

Introducing the dimensionless parameter Φ ,

$$\Phi \equiv \frac{p_0^e V_0}{H^*}, \quad (22)$$

we can rewrite equation 20 in the following form:

$$\frac{H}{H^*} = \frac{\gamma}{\gamma - 1} (1 + \Sigma) \Phi (\Theta - 1). \quad (23)$$

The dimensionless parameters Σ and Φ are not independent. Using equations 17 and 22, we arrive at the following relationship between them:

$$\Phi \Sigma^3 = \Gamma, \text{ where } \Gamma \equiv \frac{\xi^3 \sigma^3}{(p_0^e)^2 H^*}. \quad (24)$$

5. Analysis of the in-Vapor-Ignition and in-Liquid-Ignition Triggering Mechanisms

The analysis of the in-vapor-triggering mechanism is the most simple and is based solely on the analysis of equation 19. This equation shows that, at fixed parameter Θ of the required temperature raise, the parameter of Π diminishes with increase in the parameter Σ . This actually means that the smaller the initial volume V_0 of the bubble, the smaller the triggering overpressure required.

In other words, by neglecting the surface tension we get bigger values of the overpressure as compared with reality. The relative error in the estimation of the critical value of the parameter Π by neglecting the surface tension is given by the following formula:

$$\frac{\Delta \Pi}{\Pi} = \Sigma \left(1 - \frac{1}{\Theta^{1 + \frac{2}{3(\gamma-1)}}} \right). \quad (25)$$

For example, let us make the estimate $\Delta \Pi / \Pi$ taking the values for the air bubble with the radius $R_0 = 0.1$ cm within acetone at the external pressure of 1 atm = 106 dyn/cm² and the room temperature T_0 of 20 °C. The surface tension σ is equal to 25 dyn/cm. The ratio of the heat capacities $\gamma = c_p / c_v$ is approximately equal to 1.4. If for the required temperature raise $\Theta = T^* / T_0$ we accept the value of 1.1, then the formula (equation 24) gives the value $\Delta \Pi / \Pi \approx 2$. For smaller bubbles, the error can be significantly bigger.

It is clear that with such high level of discrepancy, the ignition theory without surface energy is definitely unacceptable.

6. Theoretical Analysis of the Bubble-Assisted Triggering for the van der Waals Liquid

For the realistic range of thermodynamic parameters the model of the ideal gas does not provide sufficient level of numerical accuracy or even of qualitative agreement with observations. A much better theoretical and practical agreement with existing data can be achieved by replacing the equation of state (equation 3) for the ideal gases with the equation of state of the van der Waals substances:

$$p(v, T) = \frac{RT}{v-b} - \frac{a}{v^2}, \quad (26)$$

where a and b are positive constants, depending on the substance.

The van der Waals equation of state (equation 26) does not give all the thermodynamic information about the substance. For instance, this equation doesn't anything about the specific heat of the substance. However, if the specific heat c_v at fixed volume is determined somehow, then all the thermodynamic potentials of the substance can, in principle, be derived mathematically. Quite often, the assumption of constant c_v appears to provide for a qualitatively good approximation in wide range of thermodynamic parameters. As we dwell on this simplifying assumption, it is worthy to warn of one typical mistake. It concerns the value of the heat capacity c_p at fixed pressure: for the van der Waals substance, c_p depends on the absolute temperature even if c_v does not. This follows from the identity:

$$C_p - C_v = \frac{R^2 v^3}{Rv^3 T - 2a(v-b)^2} T. \quad (27)$$

Looking closely at the analysis of the bubble-assisted triggering of explosion/combustion, one can notice that it is based on two sets of formulas. The first set (equation 12) is simply the equations expressing the Laplace formula of the tension-induced excess pressure under the curved surface. This set remains valid and unchanged for any thermodynamic model of the bubble's gas.

The second set of formulas (equation 4), on the contrary, is closely dependent upon the equation of state. For the van der Waals substance, the equation of adiabatic compression should be considerably modified. In particular, in the thermodynamic variables (v, T) the equation of adiabata takes on the following form:

$$T_0^{-\frac{C_v}{R}} (v_0 - b) = T_1^{-\frac{C_v}{R}} (v_1 - b). \quad (28)$$

The equation of adiabata in any other thermodynamic variables can be found by combining equation 28 with the equation of state (equation 26).

Thus, the quantitative and qualitative analysis of the role of surface tension in triggering explosion/combustion in non-ideal liquid substances should be based on the system of algebraic equations 11, 26, and 28.

7. Conclusion

We have re-analyzed the widely accepted theory (*I*) of initiation of explosion /combustion by means of adiabatic compression of the dissolved gaseous bubbles. The main target of our study was the role of surface tension at the interface liquid explosive/gaseous bubble. The impotence of such a study is associated with smallness of the bubbles. Their smallness causes high overpressure within the bubble as compared with pressure within the surrounding explosive/flammable liquid due to the Laplace excess pressure.

We gave the closed algebraic system of the relationships allowing one to analysis the role of the surface tension for both the ideal and non-ideal (van der Waals) gas models. The case of the ideal gas within the bubbles was explored further. Our results show the Bowden-Yoffe analysis (*I*) that neglects the surface tension always gives the smaller magnitude of power of the triggering impact. In fact, in order to initiate explosion/combustion, the impact should be bigger than the one predicted by Bowden and Yoffe (*I*). Moreover, the corrected (for the surface tension) theory raises the issue of the validity and realism of the triggering mechanism based on adiabatic compression of the dissolved gas bubbles.

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T WEERASOORIYA

INTENTIONALLY LEFT BLANK.